

Large-Scale Molecular Dynamics Simulation of Polyolefin Blends

J.G. Curro^{C, S}

Advanced Materials laboratory, Sandia National Laboratories, Albuquerque, NM, U.S.A.

E. Jaramillo

Dept. of Chemical Engineering, Colorado School of Mines, Golden, CO, U.S.A.

G.S. Grest

Sandia National Laboratories, Albuquerque, NM, U.S.A.

D.T. Wu

Dept. of Chemical Engineering, Colorado School of Mines, Golden, CO, U.S.A.

Molecular dynamics (MD) simulations were carried out on the binary blends of five polyolefins: head-to-tail isotactic (iPP) and syndiotactic (sPP) polypropylene, head-to-head polypropylene (hhPP), polyisobutylene (PIB), and polyethylene (PE). These polyolefins were modeled at the united atom level using the TraPPE potential between pairs of sites. Large systems consisting of 1600 chains of 96 sites, and 3200 chains of 48 chains, for a total of 153,600 united atoms were studied at 453K. Surprisingly, the heat of mixing for all the blends was found to depend not only on the intermolecular van der Waals contributions, but also on intramolecular van der Waals, angular bending, and torsional components. The chi parameters from the simulations were estimated from the structure factors using the random phase approximation (RPA) in analogy with neutron scattering (SANS) experiments. The MD simulations predicted chi parameters in good agreement with SANS measurements previously reported on the hhPP/PIB, hhPP/PP, and hhPP/PE blends. The simulations predicted lower critical solution temperature (LCST) behavior for hhPP/PIB, and upper critical solution temperature (UCST) phase behavior for the hhPP/PP and hhPP/PE mixtures in agreement with experiment. Intermolecular pair correlation functions were used to compare chain packing in the melts and blends. Examination of the intermolecular pair correlation functions suggests that hhPP packs more efficiently than iPP when mixed with PIB.